

Docket No.: 2224-0260PUS1
(PATENT)

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of:
Hisayoshi ITO

Application No.: 10/580,652

Confirmation No.: 5762

Filed: September 25, 2006

Art Unit: 1796

For: MULTIPLE PARTICLE AND COMPOSITION
HAVING DISPERSE SYSTEM

Examiner: G. Mesh

DECLARATION UNDER 37 CFR 1.132

MS Amendment
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

I, Mr. Hisayoshi ITO, declare the following.

I am a citizen of Japan, residing at 211-4 Oshio-cho, Himeji-city, Hyogo, Japan;

I was born on July 17, 1969 in Tokyo and graduated from Department of Molecular Chemistry and Engineering, Faculty of Engineering, Tohoku University, Aoba, Aramaki, Sendai, Miyagi, Japan in March 1994.

I have been employed by Daicel Chemical Industries, Ltd., Japan since April 1994 and engaged in Polymer Engineering. I currently work at Corporate Planning, New Business Creating Team, as a manager.

Application No.: 10/580,652

Docket No.: 2224-0260PUS1

I am the inventor of the presently claimed invention. I have read and understand the Examiner's comments in the outstanding Office Action dated July 22, 2008.

The following experiments were performed to show that all of the Inventive Examples in the present specification form particles having a core-shell structure wherein both the particle and the core thereof are spherical. The following experiments are the same experiments described in the present specification. The following experiments were either performed by me or by someone under my direct supervision:

Examples 1 to 6

In each Example, a polymer composition comprising a thermoplastic resin component and a water-soluble auxiliary component in the formulation shown in Table 1 below was melt-kneaded at a preset temperature of 200°C for 5 minutes by using a brabender (manufactured by Toyo Seiki Seisaku-sho, Ltd., laboplastmill), and then cooled to prepare a dispersion composition. The obtained dispersion composition was immersed in hot water of 60°C to give a suspension of the polymer particle. The insoluble matter was separated from the suspension with a membrane (pore size: 0.45 μm) made of a polyvinylidene fluoride to collect the fine particle of the polymer.

(Structural observation of the polymer particle)

A polymer fine particle was mixed with a chemically reactive adhesive of epoxy polymer-series (manufactured by Konishi Co., Ltd., "BONDQUICK 5") to make a massive product containing the polymer particle dispersed therein, and the massive product was cut to a thickness of about 0.05 μm to 0.2 μm by a microtome to give an ultrathin section. Thereafter, the polymer particle in the ultrathin section was stained with a coloring matter capable of staining the Polymer component (A) and the Polymer component (B) distinctively (e.g., osmic acid, and ruthenic acid), and was observed about the structure by using a transmission electron microscope.

The results of Examples 1-6 are given in the following table.

Application No.: 10/580,652

Docket No.: 2224-0260PUS1

Table 1

	Polymer (A1) [parts]	Polymer (A2) [parts]	(B1) [parts]	(B2) [parts]	Particle structure	Particle shape	Core shape
Ex. 1	Polymer 1 20	Polymer 3 20	75	25	Core-shell	Finely spherical shape	Spherical*
Ex. 2	Polymer 2 20	Polymer 3 20	75	25	Core-shell	Finely spherical shape	Spherical
Ex. 3	Polymer 2 30	Polymer 3 10	75	25	Core-shell	Finely spherical shape	Spherical*
Ex. 4	Polymer 2 20	Polymer 4 20	75	25	Core-shell	Finely spherical shape	Spherical*
Ex. 5	Polymer 1 20	Polymer 5 20	75	25	Core-shell	Finely spherical shape	Spherical*
Ex. 6	Polymer 4 20	Polymer 5 20	75	25	Core-shell	Finely spherical shape	Spherical*

(In Table, "parts" means "parts by weight")

* - Based on opinion, since transmission electron micrograph is only reported for Example 2.

(Polymer component)

Polymer 1: Polystyrene polymer (manufactured by Toyo Styrene Co., Ltd., "GPPS HRM63C")

Polymer 2: Styrene-butadiene-styrene block copolymer (SBS polymer) (manufactured by JSR Corporation, "TR2003")

Polymer 3: Polyamide 12 polymer (manufactured by Daicel-Degussa Ltd., "DIAMID L1640")

Polymer 4: Polybutylene succinate-polycaprolactone copolymer (manufactured by Daicel Chemical Industries, Ltd., "CELLGREEN CBS17X")

Polymer 5: Polylactic acid (manufactured by Mitsui Chemicals, Inc., "LACEA H-100PL")

(Water-soluble auxiliary component)

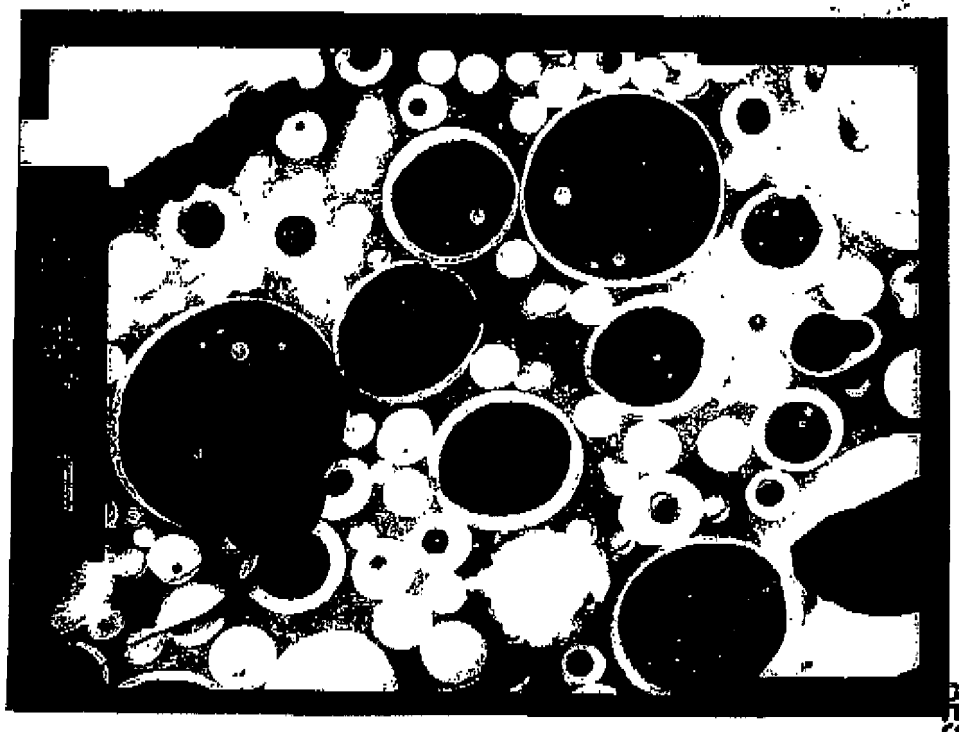
(B1) Oligosaccharide: Starch sugar (manufactured by Towa Chemical Industry Co., Ltd., reduced saccharification product of a starch "PO-10", a viscosity of a 50% by weight aqueous solution measured at 25°C by a B-type viscometer; 6.5 Pa·s)

(B2) Plasticizing component: Sugar alcohol (manufactured by Towa Chemical Industry Co., Ltd., "Marinecrystal D(-)mannitol")

Application No.: 10/580,652

Docket No.: 2224-0260PUS1

In each of Examples 1 to 6, core-shell spherically shaped polymer particles comprising the thermoplastic resin (A) as the shell were obtained. The shell thickness of each obtained particle was not more than 1 μm . In Fig. 3¹, which is reproduced hereinbelow for the reader's convenience, the transmission electron micrograph shows the cross section of the particle after staining the multiple polymer particle obtained in Example 2.



In Fig. 3, the core-shell finely spherical multiple polymer particle is shown containing the polyamide 12 polymer as the shell and the styrene-butadiene-styrene block copolymer as the nucleus (core). It was confirmed that the butadiene part of the styrene-butadiene block

¹ Using the same numbering as was used in the specification.

Application No.: 10/580,652

Docket No.: 2224-0260PUS1

copolymer forming the core was stained with osmic acid and observed black by a transmission electron microscope and that the polyamide 12 polymer formed the shell. It is clear from Fig. 3 that both the particle and the core thereof are spherical.

Moreover, it is my opinion that the samples of Examples 1 and 3-6 would also show that both the particle and the core thereof are spherical.

This structure wherein both the particle and the core thereof are spherical is the result of the method in which the core-shell particles are made. The core-shell particles of the present invention are formed by kneading a thermoplastic polymer (A1), a thermoplastic hydrophilic polymer (A2) which has a hydrophilicity higher than the polymer(A1), and a water-soluble auxiliary component (B) in a molten state, and eluting the component (B) forming the matrix of the resultant composition.

Kneading the above components in a molten state gives the composition in which the thermoplastic polymer components are dispersed in the matrix of the auxiliary component (B). On the other hand, the polymers (A1) and (A2) are different in affinity relative to the auxiliary component (B) caused by difference in hydrophilicity of the polymers (A1) and (A2). Moreover, the affinity between the polymers (A1) and (A2) is higher than their affinity relative to the component (B). Thus, when the above components are kneaded in a molten state, the polymers (A1) and (A2) together form a dispersed phase in the matrix, and the polymer (A2), which has a hydrophilicity higher than the polymer (A1), exists nearer to the component (B) than the polymer (A1) exists. That is, the dispersed phase is formed with the outer shell of the polymer (A2) and the inner core of the polymer (A1), due to the differences in their affinity relative to the component (B).

Further, in the case of an island-sea structure if both a matrix phase and a dispersed phase are in their molten states, the dispersed phase forms a spherical shape to minimize the surface tension between the two phases. This minimization of the surface tension occurs at the surface between the outer shell of the dispersed phase (the polymer (A2)) and the matrix phase (the component (B)) as well as at the surface between the outer shell of the dispersed phase (the polymer (A2)) and the inner core of the dispersed phase (the polymer (A1)). The former makes

Application No.: 10/580,652

Docket No.: 2224-0260PUS1

the shape of the entire dispersed phase spherical and the latter makes the shape of the inner core spherical. Thus, the core-shell particle itself and its inner core both have spherical shapes.

The circular shape is an inherent property of the particles which occurs when repeating the Examples of the present application. In my opinion, one of ordinary skill in the art after reading the present application would appreciate that the particle itself and its inner core both have spherical shapes because of the interactions between the dispersed phase and the matrix phase as discussed in the previous paragraph, and could confirm this by repeating the experiments described in the present application and examining the resulting particles.

In conclusion, it is my opinion that I was in possession of the invention at the instant priority date including the feature/property of the core-shell particle itself and its inner core both having spherical shapes.

I hereby declare that all statements made herein of any own knowledge are true, and that all statements made on information and belief are believed to be true; and further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Respectfully submitted,

Dated: October 21, 2008

By


Mr. Hisayoshi Ito